Response to Review Comments#1

Review of "Causes of growing middle-upper tropospheric ozone over the Northwest Pacific region" by Xiaodan Ma et al. Summary and General Comments:

This paper presents an analysis of ozone trends from ozonesonde observations and EMAC model simulations to demonstrate positive ozone trends in the mid-to-upper troposphere over the past ~30 years. The authors analyze the seasonal and spatial ozone distribution from ozonesonde profiles at Hong Kong, Naha, Tsukuba, and Sapporo and leverage model ozone tracers to attribute tropospheric ozone growth primarily to production in the troposphere, with some input from increased stratosphere-to-troposphere transport.

General Comment: The CI to ECC ozonesonde transition at the Japanese stations in ~2009 is mentioned, and the authors correctly remove the normalization factor from the ozone profile data. However, given the emphasis on ozone trends calculations, it would be prudent to demonstrate that these data do not contain a notable step-change in the time series in the Supplemental Information of this manuscript.

This paper is extremely well written, and I have only a few mostly minor suggestions, edits, and comments for the authors to address. One note: I couldn't tell if this paper has been submitted to the Tropospheric Ozone Assessment Report 2 Special Issue: https://bg.copernicus.org/articles/special_issue10_1256.html. This paper would be an excellent candidate for the issue given its topic and quality, and it may receive more attention if it is submitted to that Special Issue.

Response: Thank the reviewer for the positive feedback on the current manuscript. We added a new plot of all ozone profiles used in the analysis (Figure S1) in the P2L32 in the revised Supplementary material. After removing the normalization factors during the observation period at three Japanese sites, the corrected dataset shows no notable step-change around 2009 in Japanese sites. The related discussion has been added at P3L119-121 in the revised manuscript.
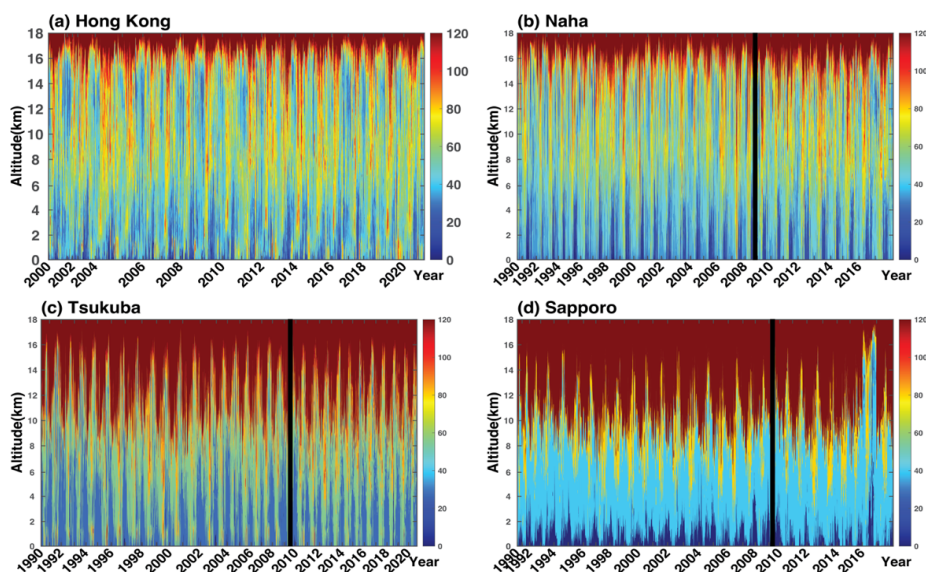

Figure S1 All O$_3$ profile samples used in the analysis at (a) Hong Kong, (b) Naha, (c) Tsukuba, and (d) Sapporo. Black lines indicate the transition time from CI to ECC ozonesonde at the Japanese stations around 2009.

Thank the reviewer for recommending the Tropospheric Ozone Assessment Report 2 Special Issue. We will apply for it after the ACPD phase.

Recommendation: I recommend this paper's publication once the authors address the mostly minor and technical comments listed below.

Specific and Line-by-Line Comments:

Line 27 (Abstract): By "hotspots" do you mean the largest trends? Reconsider the language here.

Response: Thank you for the suggestion. We intended to discuss the distinct tongue-shaped pattern in top-down direction characterized by high concentrations of O_3 extending from the lower stratosphere to the middle troposphere. It has been revised as ozone tongue in the revised manuscript.

Line 88: Because of the time response delay of the ozonesonde sensor, the profiles have at best 100 meter vertical resolution, even if the data are reported every ~5 to 10 meters.

Response: Thank you for the comment. In the sentence, we have removed the emphasis on the fine vertical resolution of less than 10 m in the revised manuscript. Please see P3L92 in the updated manuscript.

Line 117: I think you mean "underestimation" of the uncertainties rather than "overestimation."

Response: Thank you for the comment. We agree with the reviewer. The original statement has been rephrased as " the transition of the measurement technology from CI to ECC around 2009 could lead to more uncertainties and an overestimation on the long-term O_3 trends (Figure S2)." in P3L123-124. Using the corrected ozonesonde data provided by WOUDC during the CI period could lead to overestimating long-term O_3 trends (as shown below in Figure S2).
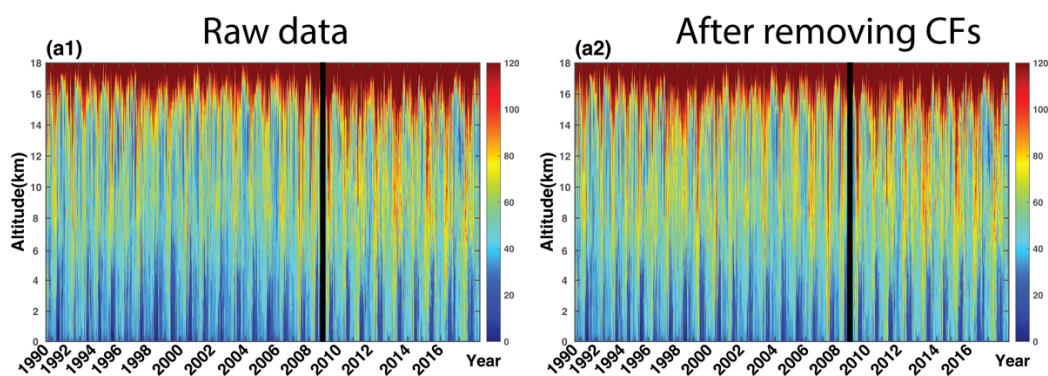

Figure S2. All O_3 profile samples were used in the analysis at Naha from (a1) WOUDC and (a2) after removing CFs. Black lines indicate the transition time from the CI to ECC ozonesonde at the Japanese stations around 2009.

Line 129: This may qualify as more than a minor comment. Both the ascent and descent data are being used here? I do have concerns about using descent profiles given the uncertainty of solution evaporation and loss from the balloon burst/tumbling through the atmosphere.

Response: Thank you for the comment. We did only use the ascent profiles for the analysis. To clarify, we modify the description as " We limit our analyses of tropospheric and lower-stratospheric O_3 profiles to altitudes below 18 km and remove duplicate O_3 values during the descent period at the same heights in the time series to prevent

redundant measurements as well as the uncertainty of solution evaporation and loss from the O_3 sounding balloon burst/tumbling through the atmosphere" in the revised manuscript P4L140-143.

Line 216: Change "like" to "such as"

Response: Thank you for the suggestion. We changed the expressions in P6L238 and P20L531 in the revised manuscript.

Line 318: ", while not so clear for the seasonal difference in the middle-upper troposphere." I don't quite follow this part. Please rewrite for more clarity.

Response: Thank you for the comment. We revised it as "This seasonal difference in the lower troposphere could be attributed to the influence of the East Asia Monsoon as discussed earlier. In the middle-upper troposphere, there are no such significant seasonal differences among sites.". Please see P11L352 in the updated version.

Line 344: This is Figure 6b3, not 7c2

Response: Thank you for the comment. It has been corrected in the revised manuscript.

Line 345: This is Figure 6a2, not 7b1. Please also correct the Figure 6 caption. For example, Hong Kong is (a1-a3), not (a1-c1).

Response: Thank you for pointing out the error. We have corrected them in the revised manuscript.

Line 356: "Figure .7", remove the period

Response: Thank you. The period has been removed.

Line 362: Change "than" to "compared to"

Response: Thank you for the suggestion. We changed it in the revised manuscript.

Figure 7: Not sure if I missed whether all model output for a station is used, or only output coincident with the ozonesonde profiles.

Response: Thank you for the comment. In this case, all the model output for a station is used. There are two considerations for using all model output rather than the coincident records. First, the monthly output of the model was then used to distinguish the ozone contribution from the troposphere and stratosphere. If the validation of monthly mean output makes sense, it will enhance the robustness of the conclusion we've reached. Second, the weekly launch frequency of the ozonesondes has been validated as reliable in representing long-term O_3 trends, as evidenced by comparing them with near-surface O_3 trends at hourly time resolution (Liao et al., 2021). Using the monthly mean of weekly resolution ozonesonde observation to validate the monthly mean of 6-hourly time resolution model output on the ozone long-term trend is feasible.

Figure 7: It would be helpful to also include some of the trend values from Table 2 on the figure itself.

Response: Thanks for the suggestion. We plotted the time series of the monthly mean of the ozonesonde observations and model output of ozone at different levels of the troposphere in Figure 7 to better compare long-term ozone changes with the seasonal variations as well as the absolute values. For a more accurate comparison

of the long-term trends, the seasonal variation should be removed. We decided to discuss the ozone trend in detail in Table 2 by separating the seasons.

Line 382: I think you mean Figure 4, not Figure 3 here.

Response: Thank the reviewer for pointing out the error. We have corrected it in the revised manuscript P14L431.

Line 521: There is an extra "u" in this sentence.

Response: Thank you. The typo has been removed.

Data Availability: Are the model output available publicly?

Response: Thank you for the question. We have uploaded the model output at Zenodo, which can be freely downloaded via https://zenodo.org/records/11093806. The related information has been updated in the revised manuscript P21L568-569.

Reference:

Liao, Z., Ling, Z., Gao, M., Sun, J., Zhao, W., Ma, P., Quan, J., and Fan, S.: Tropospheric Ozone Variability Over Hong Kong Based on Recent 20 years (2000–2019) Ozonesonde Observation, J. Geophys. Res., 126, e2020JD033054, https://doi.org/10.1029/2020JD033054, 2021.